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Screened hopping conduction in ultrathin metal films

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Abstract. We have found that screening of the Coulomb interaction in ultrathin, quench-condensed bismuth films causes the conductivity to become simply activated. We explain this in terms of fixed-range hopping of dipoles, the dipoles consisting of localized charges in the experimental film and their images in the screening electrode. We deduce localization lengths of the order of 22 nm. Non-ohmic effects imply a scale length of the order of 700 nm which probably corresponds to the typical distance between difficult hops in the critical percolation network. We predict reversion to a temperature-dependent activation energy closer to the metal–insulator transition.

1. Introduction

It is well known that the properties of a two-dimensional electron system are strongly dependent on Coulomb interaction of the conduction electrons. On the metal side of the metal–insulator transition (MIT) it results in a logarithmic quantum correction to conductivity [1] and suppression of superconductivity [2]. The interaction effects can be suppressed by the proximity of a bulk conductor at a distance of less than the characteristic scale of the interaction, believed to be the thermal length $\sqrt{D\hbar/\pi kT}$ on the metal side and the hopping distance on the insulator side.

For the weak localization regime, an estimate of how the interaction correction is reduced by the presence of bulk metal separated from the film by a layer of insulator was carried out by Altshuler *et al* [3]. A much larger effect from screening of the Coulomb interaction should take place on the insulator side of the MIT. If a metallic conductor is situated at a distance d from a two-dimensional layer with hopping conductivity, the Coulomb interaction will be screened at distances greater than d, thereby changing the layer resistance and $R(T)$ behaviour. On the insulator side of the MIT, electrons are localized and electron transport is usually by the mechanism of variable-range hopping which gives

$$\sigma(T) = \sigma_0 \exp[-(T_0/T)^p] \tag{1}$$

where, in the two-dimensional case and in the absence of Coulomb interaction, the exponent $p = 1/3$ and $T_0 \propto [g(\varepsilon_F)\xi^3]^{-1}$, $g(\varepsilon_F)$ being the density of states in the vicinity of the Fermi level ε_F and ξ is the localization radius. The Coulomb interaction here causes reconstruction of $g(\varepsilon)$: a soft linear Coulomb gap appears at ε_F: $g(\varepsilon) = g_1|\varepsilon - \varepsilon_F|$. As a result, the conductivity at low temperatures shows a different dependence with $p = 1/2$ and $T_0 \propto e^2/\epsilon_r\xi$, where ϵ_r is the relative permittivity [4].

The two-dimensional system we use in our experiments is an ultra-thin metal film in the activated region of conductivity. To detect the effect of screening more clearly,

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we compare the conductivity characteristics of such a film, deposited on metal covered with a thin insulating layer (the screened film), with those of a neighbouring film grown simultaneously on a bulk insulator (the unscreened film).

2. Experimental aspects

For our ultra-thin films we use quench-condensed bismuth. Quench condensation (deposition onto a substrate held at helium temperature) produces (disordered) films which are of relatively uniform thickness and which become continuous at very low coverage—a few monoloayers [5]. Evaporation of the bismuth was preceded by deposition of a seed film of about one monolayer of germanium as this is known to promote connectivity at low thicknesses. From earlier work [5] and from measurements with our thickness monitor, we estimate that our films became conducting at a mean thickness of about 1.5 nm. The germanium seed layer should also ensure a similar morphology for the screened and unscreened films.

Quench-condensed bismuth is a good metal with a bulk superconducting transition temperature of about 6 K, in contrast with its crystalline form, which is a semimetal. A phase transition to the crystalline state takes place if the films are warmed to 20–50 K depending on film thickness, and appears as a sharp irreversible increase in resistance. The films used in these experiments are of such high resistance (so thin) that superconductivity is totally suppressed and conduction is by hopping. The fact that the films are only structurally stable if kept at low temperatures precludes any direct structural studies.

For the screening electrode, we used an aluminium film about 100 nm thick which was allowed to oxidize in air for a couple of days to form an oxide layer about 3 nm thick [6]. It is well known that oxidation of aluminium produces a coherent oxide layer free of pinholes. The screened film was deposited over the oxidized aluminium. The oxide layers were not much above tunnelling thickness and so provided the closest possible screening electrode. We checked the insulation between the aluminium and the experimental films to ensure that any conductivity through the oxide was negligible compared with that of the samples. The unscreened film was depositied (with the germanium seed layer) close by, directly on the glass substrate.

Film resistances were measured with electrometers in the resistance range $R = 10^{12}$–10^4 Ω, and by lock-in amplifiers for $R < 10^6$ Ω. Other experimental details are given elsewhere [7], together with results obtained on the metal side of the MIT. It should be mentioned that the particular procedures we followed probably resulted in the deposition of impure bismuth films, but we do not believe that the presence of impurities in any way affects the results.

3. Experimental results

The temperature dependence of the conductivity of screened and unscreened films are shown in figure 1, where the logarithm of resistivity (resistance per square) is plotted against inverse temperature. For the unscreened film (broken curve) we see a temperature-dependent activation energy indicating variable-range hopping. Fitting the data to equation (1) in the range 2–20 K gives an excellent fit with $p \approx 0.72$ and $T_0 = 75$ K. (The data below about 1.8 K are inaccurate because those measurements were made too quickly in relation to the long time constants present in the circuit configuration used.) The value of p is sufficiently far from 0.5 to exclude straightforward variable-range hopping in an

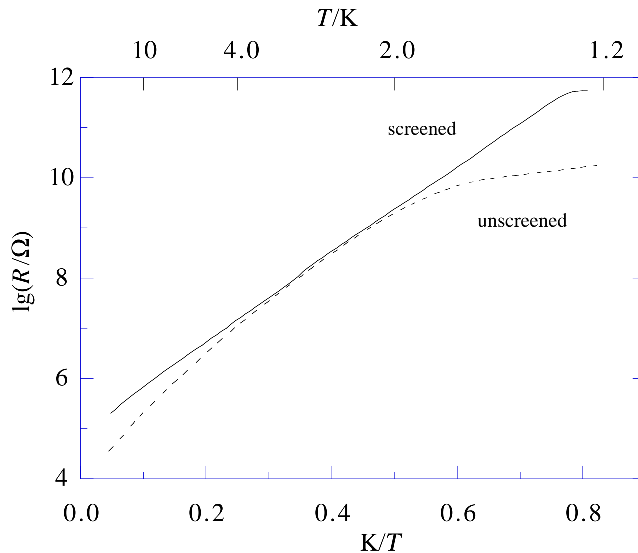

Figure 1. Logarithm of resistance as a function of inverse temperature for ultrathin Bi films. The solid curve represents results for a film deposited on an oxidized Al film (the screened film), the broken curve a film deposited directly on glass (the unscreened film).

Efros–Shklovskii Coulomb gap. There are two possible explanations.

Firstly, detailed analysis of activated conductivity in thin films shows that pure $p = 0.5$ Efros–Shklovskii behaviour is only to be expected as a low temperature limit. Above this, there can be a range where p increases, eventually rising to $p = 1$ (simple activation) when the optimum hop distance R_{opt} becomes comparable with the localization radius ξ (near-neighbour hopping). We may be in the intermediate regime. Such a scenario is consistent with the result of attempting to analyse the behaviour in terms of conventional variable-range hopping theory. The value $p = 0.72$ would imply an effective density of states rising approximately as the fourth power of $(\varepsilon - \varepsilon_F)$ and $R_{\mathrm{opt}}/\xi \approx 0.14(T_0/T)^{0.72}$. Only below 5 K is $R_{\mathrm{opt}}/\xi > 1$ implying that we have hopping to near neighbours over much of our temperature range.

The second possibility is that our films are discontinuous at these high resistances. Although discontinuous metal films typically show $p \approx 0.5$, there is evidence that p rises towards 1 in very high resistance films [8]. (It should be remembered that the actual structure could not be investigated, for example by TEM, since the films only remain structurally stable up to a few tens of kelvins.) The significance of our results is not affected by uncertainty over the explanation for the observed temperature dependence in the unscreened films since transport is by variable-range hopping in either case.

The screened results (solid line in figure 1, which incorporates several sets of data) are extraordinarily different. We observe behaviour which is very close to simple activation (Arrhenius form) over more than six orders in the resistance, with $T_0 \approx 22$ K. We also note that screening causes actual resistance values to *increase*. Increased resistances were also observed to result from screening in the metallic regime [7]. Thus in neither case does suppression of electron–electron interaction increase conductivity as might have been expected. We also noticed that conduction far more easily became non-ohmic in the screened films. We discuss this further in the next section.

4. Discussion

The central result of our experiments is that screening of the Coulomb interaction causes hopping conductivity to become simply activated. It is relevant that, some years ago, Entin-Wohlman and Ovadyahu found similar behaviour in experiments with a form of indium oxide [9]. Their results with that system differed from ours in that both resistance and activation energy were increased at all measurement temperatures. However, their observation of a change to Arrhenius behviour in a very different hopping system does suggest that screening of the Coulomb interaction will cause a change to simple activation in any hopping system otherwise showing Coulomb gap behaviour. In other words, our central result is general and not specific to the particular system we have studied.

In our experimental configuration, screening of the interaction between charges only becomes effective at distances significantly greater than the thickness d of the insulator layer between the experimental film and the screening electrode. Clearly, a systematic study of conductivity as a function of insulator thickness would give a direct measure of the optimum hop distance in the variable-range regime. This would be a valuable investigation but we have not yet been able to carry it out.

The observation of Arrhenius behaviour over such a large range of resistance makes it extremely unlikely that we are observing some chance compensation between different processes which happens to give such an overall behaviour. There must be a single, well defined activation energy that controls conductivity and we must seek an explanation in such terms.

An obvious, well defined energy of the screened system is the electrostatic energy of a charge and its image in the screening electrode. We expect the localization length ξ of the localized charges to be much greater than the dielectric thickness d, so the configuration approximates a parallel-plate capacitor with area $\pi\xi^2$, electrode separation d and dielectric of relative permittivity 8.5 (alumina). Equating the capacitative and activation energies gives $\xi \approx 22$ nm, a perfectly reasonable value. It should be noted here that there is no reason why the localization length should be the same in unscreened and screened films. In fact, quite the reverse is to be expected. We observe that the conduction process is changed profoundly by screening, so there is every reason to expect ξ to be changed too. If the capacitative energy were totally dominant, ξ would become as large as possible to maximize the capacitance and minimize the electrostatic energy. Clearly, it is the disorder energies that prevent this from happening and set the actual value of ξ.

How could the electrostatic energy be the characteristic energy involved in hopping of carriers? The situation is superficially reminiscent of polaron hopping where the energy of a localized carrier is lowered by relaxation of surrounding ions; hopping to a new site then occurs so rapidly that there is insufficient time for the ions to respond during the charge transfer and the energy originally gained by relaxaton has to be supplied for a hopping transition to occur. This situation arises because the ions cannot react more rapidly than an inverse phonon frequency, which is greater than the tunnelling time. A similar inequality seems unlikely in our system since the screening is electronic and we would expect the response to be of the order of the plasmon frequency of the screening electrode. This expectation is confirmed by two simple model calculations. Firstly, we suppose that the localized charge is transferred to a new site instantaneously and ask how quickly the screening charge can move to its new site. The situation is represented by a simple R–C circuit in which R is the resistance presented by the skin effect in the surface of the screening electrode. If the charge moves a distance of several ξ, the relaxation time comes out to be of the order of 10^{-15} s. Secondly, we may ask how rapidly the electric

field can propagate to the new site, treating the structure as a waveguide. This gives a similar time. These estimates have to be compared with the tunnelling time. Tunnelling times remain controversial, but we may identify a lower limit by applying the uncertainty principle to the fact that we measure a well defined activation energy ($= kT_0$). This gives a tunnelling time of $> 4 \times 10^{-13}$ s, which is considerably greater than the relaxation time for the screening charge. We conclude that the localized charge and its image move as an entity.

This conclusion is consistent with the changed temperature dependence of the conductivity. If the screening charge were left behind, the electrostatics of hopping would be no different from the normal Efros–Shklovskii situation with a long-range Coulomb interaction, and the usual $p = 0.5$ dependence would be expected. We therefore require an explanation for the observed conductivity in terms of *hopping of dipoles*.

We may also deduce that conduction does *not* involve variable-range hopping. This may be seen as follows. In variable-range hopping we optimize the two exponential terms describing the hopping rate in the basic expression for the conductivity:

$$\sigma = \sigma_0 \exp -(W/kT + 2R/\xi) \tag{2}$$

where W is the hopping energy and R the tunnelling distance. To obtain an overall behaviour which is simple activation, R_{opt} must be temperature independent. We therefore conclude that the tunnelling distance is not (significantly) dependent on temperature.

The basic treatment of Efros–Shklovskii hopping simply takes as dominant the energy required to separate a hopping charge carrier from the 'hole' it leaves behind. Stability of the ground state then requires the Coulomb gap and gives the standard $p = 0.5$ behaviour. This dependence is consequent on the Coulomb interaction being long range ($\sim 1/r$). In our screened system the corresponding energy would be that required to separate a dipole from the (dipole) hole it leaves behind. The mutual potential energy is now short range, varying as $1/r^3$. The consequent behaviour will depend on the hop distance, R_{hop}. If this is short enough that the variation with distance of the dipole–dipole potential still dominates the energetics, one would have to have variable-range hopping. This is clearly not the case in our results. If we have $R_{\text{hop}} > \xi$, the variation with distance of the dipole–dipole energy will rapidly become negligible in comparison with variations of potential from disorder. Tunnelling is then simply to the nearest available site and there is no optimization of the two terms in equation (2). We therefore suggest that we have fixed-range hopping of dipoles over distances $R_{\text{hop}} > \xi$. Identification of the activation energy with the electrostatic energy of the dipoles follows from this inequality since, before the hop, we have one dipole and, after the hop, we have two independent dipoles, the transferred dipole and the (dipole) 'hole' left behind. The energy to create a dipole is thus the characteristic energy of the hopping process.

We should emphasize the relation with background disorder. The electrostatic energy of our dipoles is about 2 meV. Disorder potentials will be considerably greater than this (equal to the binding energy of a trapped carrier with two-dimensional Bohr radius $\approx \xi$). The ground state of the system will therefore have many trapped dipoles of both signs. The presence of many dipoles in the ground state is analagous to the presence of many charged grains in the ground state of granular metals [10].

Finally, we return to the non-ohmic behaviour of the screened films. We have not studied this in detail, but we have made enough measurements to extract another characteristic length. If we assume the field modifies the forward and reverse hopping probabilities of the dipoles by lowering and raising respectively the energy required for transitions, we obtain

$$I \propto \exp -(W_0/kT)\,\{\exp(x) - \exp(-x)\}$$

where W_0 is the basic activation energy, and $x = eEL/kT$ is the amount by which forward/reverse energies are lowered/raised. E is the applied field in the experimental film and L the characteristic length. At constant temperature, we therefore have

$$-\ln R \propto \ln(\sinh x/x).$$

Comparing resistance values at two different applied voltages we obtain corresponding values of x and find that they are roughly linear in $1/T$ as required by this treatment (figure 2). The gradient gives $L \approx 700$ nm. This must be either a typical hop distance or the distance between difficult hops in the critical percolation network. Since L/ξ is so large (about 20), the latter seems the more likely. These figures appear satisfactory.

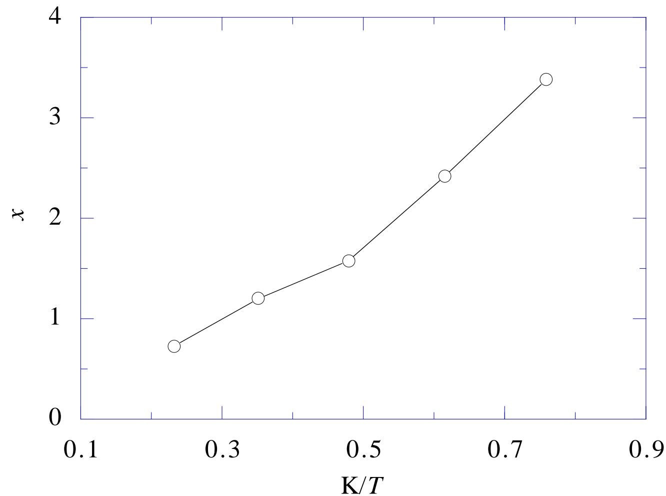

Figure 2. Variation of the non-ohmic parameter x with inverse temperature.

5. Summary

We have found that screening of the electron–electron interaction in ultra-thin metal films causes the hopping conduction to become simply activated. We explain this in terms of the hopping of dipoles, each dipole consisting of a localized charge in the experimental film and its image in the screening electrode. We have shown that these dipoles hop as an entity. The activation energy is identified with the electrostatic energy required to create these dipoles. When the hop distance is beyond the range of the dipole–dipole interaction, the activation energy becomes (essentially) independent of distance and the hop is (essentially) fixed-range, with the range being determined by the density of localized states available. This is our explanation for the Arrhenius behaviour observed in our experiments and in those of Entin-Wohlman and Ovadyahu [9]. On the basis of our model, we predict that higher densities of localized states would lead to shorter hopping distances and a regime in which there will be variable-range hopping and a temperature-dependent activation energy.

Acknowledgments

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